The Effect of Non-Linearity on a Two-Dimensional Model of Crystal-Growth Disorder

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(Received 11 April 1975; accepted 26 June 1975)

Some mathematical and optical-analogue results are used to describe the effect of introducing a non-linear term into the regression equation defining a previously described model of crystal-growth disorder. With this extended model it has been found that lattices may be produced which have completely different distributions of the two scattering species but which nevertheless give rise to indistinguishable diffraction patterns. In addition it has been found that in certain cases the disorder diffuse scattering has two maxima within the reciprocal unit cell even though only nearest-neighbour interactions are involved.

I. General introduction

In a previous paper (Welberry & Galbraith, 1973) a two-dimensional model of crystal-growth disorder was described and results for a special case (obtained by imposing a restriction on the model) for which a mathematical solution was possible, were presented. An optical-diffraction analogue technique using computer-simulated realizations of the model was also described and this technique used to verify and illustrate the mathematical findings. It was evident from this work that while the restriction made resulted in a very special case from a mathematical viewpoint there was no reason to suppose that it should represent a realistic simplification in practical terms. The present paper attempts, by means of presenting some further mathematical results and optical simulations, to describe the effect on this simple short-range-order system when the restriction is removed, although as yet a full mathematical solution has not been found.

II. Background

The disordered lattice is represented by an array of (0,1) random variables \( x_{i,j} \) and is produced by means of the algorithm:

\[
P(x_{i,j} = 1 | x_{i-1,j}, x_{i,j-1}) = \alpha + \beta x_{i-1,j} + \gamma x_{i,j-1} + \delta x_{i,j-1} x_{i-1,j} \tag{1}
\]

after the boundaries \( x_{0,j} \) and \( x_{i,0} \) have been arbitrarily assigned. In the optical simulation masks a hole is taken to represent \( x_{i,j} = 1 \) and a blank to represent \( x_{i,j} = 0 \).

The restriction that was made in the work described previously (Welberry & Galbraith, 1973), namely \( \delta = 0 \), makes equation (1) linear and in this form the equation was solved and expressions for the concentration of 1's, \( \theta \), and the general two-point correlation coefficient, \( \rho_{r,s} \), were obtained. For this simplified version of the model it was found that the three independent starting parameters \( \alpha, \beta, \gamma \) gave rise to three independent measurable quantities; \( \theta \), the concentration, and \( \rho_{0,1}, \rho_{1,0} \) the first-order correlation coefficients in the \([0,1]\) and \([1,0]\) crystal directions. By varying \( \alpha, \beta, \gamma \) this linear version of the model enabled realizations to be produced with \( \rho_{1,0}, \rho_{0,1} \) having independently any value over their whole \(-1\) to \(+1\) range and, for each of these, \( \theta \) could take any value over a range also. The question arose, therefore, as to whether the non-linear term \( \delta \) could usefully be considered as a further degree of freedom by which a lattice having a given combination of \( \theta, \rho_{1,0}, \rho_{0,1} \) could vary, and if so in what way this was most obviously manifest in the diffraction pattern. This line of approach has been adopted in the following description.

III. Present investigation

The present paper is concerned with the cases \( \delta \neq 0 \). Some results can immediately be obtained from equation (1). If expectation values are taken (1) becomes

\[
\theta = \alpha + (\beta + \delta)\theta + \delta P_{1,T} \tag{2}
\]

Secondly, with equation (1) multiplied by \( x_{i-1,j-1} \) and expectations taken,

\[
P_{0,1} = (\alpha + \gamma)\theta + (\beta + \delta)P_{1,T} \tag{3}
\]

and similarly with a multiplying factor of \( x_{i,j-1} \),

\[
P_{0,1} = (\alpha + \gamma)\theta + (\beta + \delta)P_{1,T} \tag{4}
\]

where \( P_{1,T}, P_{0,1}, \) and \( P_{1,0} \) are joint probabilities of two 1's occurring as neighbours in the \([1,1]\), \([0,1]\), \([1,0]\) crystal directions, and are related to the corresponding correlation coefficients by,

\[
\rho_{r,s} = \frac{P_{r,s} - \theta^2}{\theta(1 - \theta)} \tag{5}
\]

The three equations (2)-(4) represent necessary restrictions on the concentration and correlations that are produced in a realization of the model but do
not provide sufficient information for their determination. One additional piece of information is required to achieve this. In the next section two special (though non-linear) cases are described for which $\theta$ has been determined and hence equations (2)–(4) solved to obtain the first-order correlations.

IV. Determination of concentration and low-order correlations

The two special cases for which $\theta$ has been determined are best described in terms of an alternative formulation of the growth process.

Suppose, when a realization of the model is produced, the row $j$ is to be added to a crystal in which the row $j-1$ is complete. The points $i=1, 2, 3, ...$ etc. are filled in according to equation (1). The transition from the point $i-1$ to $i$ in this $j$th row is thus effectively produced by means of one or other of the transition probability matrices derived from equation (1),

$$B_0 = \begin{pmatrix} 1 - \alpha & 1 \\ \alpha - \beta & \alpha + \beta \end{pmatrix}$$

$$B_1 = \begin{pmatrix} 1 - \alpha - \gamma & \alpha + \gamma \\ \alpha - \beta - \gamma - \delta & \alpha + \beta + \gamma + \delta \end{pmatrix}$$

If $x_{i,j-1} = 0$ then $B_0$ is used to obtain the value of $x_{i,j}$ and if $x_{i,j-1} = 1$ then $B_1$ is used. The distribution of $x_{i,j}$ is then given by a product of $i$ such matrices ($B_0, B_1$), the order of multiplying being determined by the realization of the previous $(j-1)$th row. For example the realization 00101011 ... in row $j-1$ gives rise to the distribution $B_0 B_0 B_1 B_0 B_1 B_0 B_1 ...$ in row $j$.

This analysis yields explicit answers in the following two special cases.

(i) Special case 1 (hereafter referred to as SC1), $B_0 B_1 = B_1 B_0$

If the matrices $B_0$ and $B_1$ commute, then the order of multiplication is immaterial and the distribution of $x_{i,j}$ is only dependent on the proportion of 0's and 1's in the previous row and not on the order in which they arise. The restriction on the starting parameters necessary for the matrices $B_0$ and $B_1$ to commute is

$$(\alpha + \gamma) (1 - \alpha - \beta) = \alpha (1 - \alpha - \beta - \gamma - \delta)$$

i.e.

$$\gamma (1 - \beta) = -\alpha \delta.$$  \hspace{1cm} (6)

In this case the sequence of matrix products collapses to $B_0^r B_1^r$ where $r$ is the number of 1's in the first $i$ places of the previous row and, if the process is stationary, the concentration may be obtained as

$$E(x_{i,j}) = \lim_{r,s \to \infty} B_0^s B_1^r = \frac{\alpha}{1 - \beta}$$

i.e.

$$\theta = \frac{\alpha}{1 - \beta}.$$  \hspace{1cm} (7)

If this result is combined with equations (2)–(4) and (6),

$$P_{1,1} = \frac{\alpha^2}{(1 - \beta)^2} = \theta^2$$

$$P_{0,1} = (\alpha + \gamma) \theta + (\beta - \delta) \theta^2 = \frac{\alpha^2}{(1 - \beta)^2} = \theta^2$$

$$P_{1,0} = (\alpha + \beta) \theta + (\gamma + \delta) \theta^2$$

and from these probabilities the corresponding correlation coefficients are, by equation (5),

$$\varrho_{1,1} = \varrho_{0,1} = 0$$

$$\varrho_{1,0} = \beta - \gamma.$$  \hspace{1cm} (9)

(ii) Special case 2 (hereafter referred to as SC2) $B_1 = B_1^t = B_0 B_1$

There are two conditions necessary for this property to hold;

$$\gamma = 0 \quad \text{and} \quad \beta = -\delta.$$  \hspace{1cm} (10)

In this case the sequence of matrix products collapses to $B_1^r B_0^r$ where $r$ is the number of 0's before the first 1 in the previous row. If the lattice is stationary the probability that $r$ takes a certain value in a given row is independent of the previous row (since $\gamma = 0$). Thus,

$$P(r=0) = \theta$$

$$P(r=j) = (1 - \theta) \alpha (1 - \alpha)^{j-1} \quad \text{for} \quad j = 1, 2, 3, ... \text{etc.}$$

and the following results may be obtained, (see Galbraith, to be published)

$$\theta = \frac{\alpha(1 + \alpha \beta)}{1 - \beta + 2 \alpha \beta}$$  \hspace{1cm} (11)

$$P_{1,1} = \theta \cdot \frac{2 \alpha (1 + \beta)}{1 + \alpha \beta} = \theta^2 - (\alpha - \theta)^2$$

$$P_{1,0} = \theta \cdot \left[ \frac{1 + \beta (1 - \alpha)}{1 + \alpha \beta} \right] = \theta - \alpha (1 - \theta)$$

whence the correlations are,

$$\varrho_{1,1} = \frac{(\alpha - \theta)^2}{(1 - \theta)}$$  \hspace{1cm} (12)

$$\varrho_{1,0} = \frac{\theta - \alpha}{\theta}$$  \hspace{1cm} (13)

$$\varrho_{0,1} = \frac{\alpha - \theta}{1 - \theta}.$$  \hspace{1cm} (14)

There are two other related special cases that are obtained in the same way as for SC1 and SC2 when growth is considered to proceed along rows of constant $i$.

It is also worth stressing at this point that although the above results have been obtained for SC1 and SC2 these do not include any knowledge of the behaviour of other higher-order correlation coefficients and in particular the correlation $\varrho_{1,1}$ in the growth direction.
V. Experimental investigation of SC1 and SC2

The two special cases described in the last section have been studied by the optical-analogue technique described by Welberry & Galbraith (1973) and the more recent development of the method using an Optronics Photowrite System (see Harburn, Miller & Welberry, 1974). It was a prime aim of the investigation to see how a lattice having particular values of \( \theta, \varrho_{1.0}, \) and \( \varrho_{0.1} \) could vary with the introduction of the non-linear term \( \delta \).

From this point of view SC1 provides the most interesting study since the starting parameters \( \alpha, \beta, \gamma, \delta \) are subject to only one restraint, namely equation (6). According to equation (9) \( \varrho_{0.1} \) is fixed at zero so only \( \theta \) and \( \varrho_{1.0} \) may be chosen independently, and when this has been done one other variable can be chosen (e.g. \( \delta \)). As an illustration of this series \( \theta \) was fixed at 0.5 (since this gives optimum diffuse diffraction) and simulations performed for different values of \( \varrho_{1.0} \) and \( \delta \). Table 1 lists values of the starting parameters necessary for this survey. Fig. 1(a) shows representative small portions of the realizations and Fig. 1(b) their corresponding diffraction patterns. Only those realizations given in Table 1 which have \( \delta \) negative are illustrated in Fig. 1 since those with \( \delta \) positive may be obtained from those with \( \delta \) negative merely by interchanging 0's and 1's in the arrays (see § V1.iv). Since for this series neither the concentration (= 0.5) nor the two-point correlations would be affected by such a change neither would the diffraction pattern.

SC2, being the result of imposing two restraints on the starting probabilities is less general and if \( \theta \) and \( \varrho_{1.0} \) in equations (11) and (13) are chosen independently the starting parameters are then fixed uniquely. It is found however that \( \varrho_{1.0} \) may not be assigned any

Table 1. Values of the starting parameters \( \alpha, \beta, \gamma, \delta \) for the series SC1 which give rise to realizations with the particular values of \( \varrho_{1.0} \) given in the first column and a concentration of 0.5

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<th>( \alpha )</th>
<th>( \beta )</th>
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arbitrary value since only lattices with moderately low values of $|\varphi_{1,0}|$ are possible. For example, with $\theta = 0.5$ the maximum permissible value of $\varphi_{1,0}$ is 0.414.

To illustrate this series $\theta$ was again fixed at 0.5 and lattices with various values of $\varphi_{1,0}$ produced. The values of the starting parameters necessary for this series are given in Table 2 and Fig. 2 shows portions of the realizations and their diffraction patterns. Included also in Table 2 and Fig. 2 are the corresponding linear examples which have the same values of $\theta$, $\varphi_{1,0}$, $\varphi_{0,1}$ as each of the SC2 examples. Note that there is a complementary series to SC2 (for which $\beta = 0$; $\gamma = -\delta$) which gives rise to the same series of diffraction patterns as SC2. The lattices of this second series may be obtained from those of SC2 by interchanging 0's and 1's.

VI. Some properties of the non-linear model

(i) Degree of non-linearity

It has been found possible for SC1, as described in the previous section, to produce a range of lattices having a particular combination of $\theta$, $\varphi_{1,0}$, $\varphi_{0,1}$, the quantity variable over the range being the non-linear term $\delta$. The range of $\delta$ permissible for a given combination depends on their particular values. In general, high values (positive or negative) of the correlation coefficients $\varphi_{1,0}$, $\varphi_{0,1}$ permit only a small variation in $\delta$.

Table 2. Values of the starting parameters $\alpha$, $\beta$, $\gamma$, $\delta$ for the series SC2 which give rise to realizations with the particular values of $\varphi_{1,0}$ and $\varphi_{0,1}$ given in the first two columns and a concentration of 0.5

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<th>$\varphi_{0,1}$</th>
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Fig. 1. (a) Diffraction masks of SCI produced with the starting parameters given in Table 1. The areas shown are small representative portions of the much larger arrays that were used to obtain the diffraction patterns shown in Fig. 1(b).
Fig. 1 (cont. (b)) Diffraction patterns of SC1. Each pattern was obtained by diffraction from a circular area of the mask containing about 200,000 lattice points.
whereas low correlations permit a considerable variation (see Table 1 and Fig. 1). The very special case \( \varrho_{1,0} = \varrho_{0,1} = 0 \) permits a variation of \( \delta \) over the whole \(-2 \) to \(+2\) range. Values of \( \delta \) outside the ranges given result in at least one of the transition probabilities taking values outside the \( 0-1 \) range.

For a given choice of \( \theta, \varrho_{1,0} \) SC2 gives rise to a single lattice with a unique value of \( \delta \) (see Table 2). This does not however mean that this is the only value of \( \delta \) permissible for this particular combination of \( \theta, \varrho_{1,0}, \varrho_{0,1} \), but that only this value corresponds to SC2. There is a complete range of structures possible extending in a similar manner to SC1 from the linear case to a maximum \( \delta \) case, the SC2 case being a single example in the range. Only for the linear case and the SC2 case has it been possible to determine \( \theta \), and hence obtain the starting parameters \( \alpha, \beta, \gamma, \delta \). The first and last examples in Table 2 are ones in which the SC2 case is also the maximum permissible \( \delta \) case.

Since for each combination of \( \theta, \varrho_{1,0}, \varrho_{0,1} \) which is possible in SC2 the corresponding linear example is determinable, it has been possible to estimate other members of the series with different values of \( \delta \) by interpolation or extrapolation. It has been found that linear interpolation enables sets of \( \alpha, \beta, \gamma, \delta \) to be derived which are close approximations to other members of the series, i.e., these interpolated values give rise to lattices in which the observed values of \( \theta, \varrho_{1,0}, \varrho_{0,1} \) are very close to the values for the linear and SC2 cases. However, although the agreement is very close, some significant differences are suspected and this work is still in progress. 

(ii) Manifestation of the degree of non-linearity in the diffraction patterns

For a given set of \( \theta, \varrho_{1,0}, \varrho_{0,1} \) the variation in the degree of non-linearity produces in general a corresponding variation in the diffraction pattern [with one notable exception; see (iii) below]. The question arose therefore as to what feature of the diffraction patterns could best be used to categorize the degree of non-linearity. Since measurement of the whole 2-D field of correlation coefficients is not a feasible proposition, attention was given to the higher-order correlations in the \( [0,1] \) and \( [1,0] \) axial directions which are in principle obtainable from the diffuse peak profiles in these directions. Tables 1 and 2 include estimates of some of these higher-order correlations which were obtained by counting on generation in the computer. Note that \( C_{r,s} \) is taken to mean the experimental estimate of \( q_{r,s} \).

With reference to Fig. 1, for SC1 the diffuse peak profiles are readily related to the behaviour of these higher-order correlations. It is noticed in Table 1 that in general with increase in \( |\delta| \) for examples with negative \( \varrho_{1,0} \) the values of \( C_{2,0}, C_{3,0}, C_{4,0} \) are numerically less than their corresponding values for the linear (\( \delta = 0 \)) case and examples with positive \( \varrho_{1,0} \) have \( C_{2,0}, C_{3,0}, C_{4,0} \) greater than their corresponding values for the linear case. The effect of these changes on the diffraction peak profile must be studied in detail for individual

![Fig. 2. Diffraction masks and patterns of SC2 compared with those for linear examples having the same \( \theta, \varrho_{1,0}, \varrho_{0,1} \). (See Table 2 for details of the starting parameters.) The regions of the masks illustrated are small representative portions of the much larger arrays used to obtain the diffraction patterns, as in Fig. 1.](image)

![Fig. 3. Diffuse peak profiles for some examples of SC1. In each of the four series of constant \( \varrho_{1,0} \), the upper trace is of the linear (\( \varrho = 0 \)) example and the lowest of the most non-linear. The traces were obtained by scanning the films used to produce Fig. 1(b) in a horizontal direction (i.e., normal to the diffuse bands) with a photometer. The scanning slit was narrow in the direction of scan and wide in the direction normal to this, almost, but not quite, extending over one reciprocal unit cell.](image)
cases but in general the effect of increase in non-linearity is such that for positive $q_{1,0}$ the diffuse peak becomes narrower and for negative $q_{1,0}$ broader. Fig. 3 shows photometer scans of the profiles for the series when $q_{1,0} = -0.4, -0.2, +0.2, +0.4$.

Two particular patterns are worth a special mention. These are those for which $\delta = -1.6$ and $q_{1,0} = \pm 0.2$. These two patterns show two diffuse peaks within the reciprocal unit cell. They are most remarkable since they indicate most clearly stronger interactions between distant neighbours than would be expected according to previous theories when only nearest-neighbour interactions have been introduced at growth (Welberry, 1975).

Although the broadening or narrowing effect described above is not visually so obvious in the patterns of SC2 (see Fig. 2) reference to the observed correlation coefficients reveals that very similar effects are present for these examples. For example compare the case $\delta = 0.6897$, $q_{1,0} = -0.4$ from Table 2 with the same case obtained from Table 1 by interpolation (Table 3). This would suggest that to a first approximation at least the higher-order correlations are dependent only upon the first-order correlation in that direction and the degree of non-linearity and not on the value of correlations in the other axial direction.

Table 3. Comparison of results from Tables 1 and 2 for $\delta = 0.6897, q_{1,0} = -0.4$

<table>
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<th>C₁,₀</th>
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<th>C₃,₀</th>
<th>C₄,₀</th>
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</table>

(iii) Special case of zero correlations

Although the condition $q_{1,0} = q_{0,1} = 0$ has not been proved to imply that all non-origin correlation coefficients must be zero, both the appearance of the diffraction patterns (see Fig. 1) and the results obtained by counting indicate that within the error expected from the finite sample of the arrays (500 x 500 points) this is in fact the case. This means that the diffraction patterns for all members of this series are indistinguishable unless very small samples are taken. It is therefore not possible merely by observing the diffraction pattern of one of these examples to give any indication of the degree of non-linearity. The importance of this fact has been stressed by Taylor & Welberry (1974).

In contrast, the appearance of the lattices for this series does vary widely from the purely random ($\delta = 0$) case to the quite highly ordered ($\delta = -2.0$) non-linear case. If such distributions as these existed in real crystals there is little doubt that their general physical properties would be different even though they would be indistinguishable from diffraction evidence alone.

(iv) Homometric pairs of lattices

Since interchanging 0's and 1's in any realization of the model does not affect the two-point correlation coefficients, for realizations for which $\theta = 0.5$ such a transformation will not affect the diffracted intensity distribution. The pairs of lattices produced by such a transformation may then be called homometric (Patterson, 1944). If $\theta \neq 0.5$ pairs of lattices giving the same diffuse intensity but different lattice peak intensities are produced.

It may readily be shown that interchanging 0's and 1's is equivalent to a transformation from one member of the model with starting parameters $\alpha$, $\beta$, $\gamma$, $\delta$ to another member with parameters $\alpha_1$, $\beta_1$, $\gamma_1$, $\delta_1$, where,

\[
\begin{align*}
\alpha_1 &= 1 - \alpha - \beta - \gamma - \delta \\
\beta_1 &= \beta + \delta \\
\gamma_1 &= \gamma + \delta \\
\delta_1 &= -\delta.
\end{align*}
\]  

(15)

Two examples of this property are given in Fig. 4. It is noticed that while the non-linear pair are readily seen to be complementary and their textural appearances to be quite different, the linear pair have very similar textural appearances, disguising the fact that they are in fact complementary. This is explained by reference to equations (15) since for $\theta = 0.5$ a linear example transforms into itself. [Note that when the two arrays are produced with sequences of pseudo-random numbers $R_i$ and $S_i$, for the two realizations to be exactly complementary $R_i$ must equal $(1-S_i)$ for all $i$.]

![Fig. 4. Homometric pairs of lattices. The four lattices shown are realizations of: (a) SC1 with $q_{1,0} = 0.4$; $\delta = +1.2$; (b) SC1 with $q_{1,0} = 0.4$; $\delta = -1.2$ produced with the complementary sequences of pseudo-random numbers to that of (a); (c) SC1 with $q_{1,0} = 0.4$; $\delta = 0$, i.e. linear; (d) Same as (c) but using complementary sequence of pseudo-random numbers.](image-url)
This property emphasizes a basic difference between linear and non-linear examples. Whereas in linear lattices the distributions of the two species (0 and 1) are the same (if $\theta = 0.5$), in highly non-linear lattices their distributions are quite different. If a real mixed crystal containing equal proportions of two different species had grown according to the present model and gave a diffraction pattern corresponding to the example in Fig. 4 [see Fig. 1(b)], it would be impossible to say which of the homometric pair had grown unless additional non-diffraction evidence were available.

(v) Independence of lattice rows
Another aspect of the SC1 examples is of interest. While it is found both from the appearance of the diffraction patterns and the view of the correlation field found by counting that there is no correlation between a point in one row with any single point in the previous row (above), it is evident in the more non-linear examples that adjacent horizontal rows are by no means independent. For example, the presence in some realizations of the large blank triangular regions means that for two rows passing through this area a long string of blanks occurs at practically the same position. In this case with the absence of any two-point correlations it is the three-point correlations (relating a point in one row jointly with two in another) that maintain the dependence of one row on the next. This dependence evidently decreases as the linear model is approached and in fact the linear examples of SC1 do consist of independent Markov chains running horizontally.

VII. Conclusion
Although the two special cases described in this paper do not provide a comprehensive survey of the non-linear model, their investigation has pin-pointed several significant features of two-dimensional disorder that were previously unsuspected. Firstly, the starting parameters $\alpha$, $\beta$, $\gamma$, $\delta$ are no longer always uniquely determinable from the diffraction pattern. In particular, when all two-point correlation coefficients are zero, a whole range of different lattice distributions is possible which gives rise to a series of indistinguishable diffraction patterns. Secondly, a key difference between the types of lattice which are produced by the linear and non-linear model appears to be that, while for linear examples the distributions of the two species (hole and blank) are similar, for non-linear examples their distributions are quite different. Finally, from a diffraction viewpoint, the most significant finding is that the introduction of non-linear effects can result in the appearance of more than one diffuse peak within the reciprocal unit cell – such an effect would previously have been thought to have required direct interaction between neighbours of two or more units separation.

As mentioned before the linearity condition previously imposed on the model seems unlikely to be a reasonable physical condition for real crystal growth. It seems likely, therefore, to be more rewarding to undertake studies of this and other non-linear systems rather than to extend the present model to other linear models which include interactions between more neighbours.

References